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(54) Process for the preparation of plastoelastomeric compositions based on a thermoplastic olefinic polymer

(57) Process for the preparation of plastoelastomeric preparations composed by an elastomeric phase constituted by EPDM elastomers and by a plastomeric phase constituted by thermoplastic olefinic polymers, wherein said elastomeric phase is dynamically vulcanized with a vulcanizing binary agent consisting of an alkylphenol-formaldehydic resin and sodium bisulphite.

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PROCESS FOR THE PREPARATION OF PLASTOELASTOMERIC COMPOSITIONS  
BASED ON A THERMOPLASTIC OLEFINIC POLYMER

This invention refers to a process for the preparation of plastoelastomeric compositions based on a thermoplastic olefinic polymer, formed by an elastomeric phase constituted by a thermoplastic olefinic polymer, wherein said elastomeric phase is dynamically vulcanized.

More in detail, this invention refers to a process for the preparation of plastoelastomeric compositions.

The invention also refers to plastoelastomeric compositions obtainable with said process as well as to the artifacts obtained from them.

The process of dynamic vulcanization is well known in the art and described, for instance, in the Italian patent nr. 948902.

Such patent describes the preparation of plastoelastomeric compositions whose elastomeric phase is dynamically vulcanized by means of organic peroxides.

More systems of dynamic vulcanization of the elastomeric phase of plastoelastomeric compositions are described in US patents nr. 3 997 487, 4 247 652, 4 409 365 and 4 835 204.

However, the plastoelastomeric obtained through the known vulcanization systems have physical-mechanical and processability properties that are not entirely satisfactory, which cause the compositions to be unsuitable for all the applications for which they should be utilized.

Object of this invention is to solve this drawback of the known vulcanization systems.

More in detail, object of this invention is to provide a

vulcanization system such as to permit the preparation of dynamically vulcanized plastosoluble compositions having improved physical-mechanical and processability properties.

According to this invention, these and still other objects are achieved by using a binary vulcanization system constituted by an alkylphenol-formaldehydic resin and sodium bisulphite.

The object of this invention is therefore a process for the preparation of plastoelastomeric compositions formed by a plastomeric phase consisting of a thermoplastic olefinic polymer and a dynamically vulcanized elastomeric phase wherein dynamic vulcanization has been obtained by using a binary vulcanizing system consisting of an alkylphenol-formaldehydic resin and sodium bisulphite.

The plastoelastomeric compositions of this invention are obtained by submitting to mastication a mix of an elastomer (elastomeric phase) and a polyolefin (plastomeric phase), in the wished weight rates, at temperatures high enough to permit the fusion of the plastomeric phase, for instance from 150 to 190°C, according to the type of polyolefinic elastomer, and for a time long enough to obtain an homogeneous mixing of the two components. To this purpose, one can use the common apparatuses and technologies known and utilized in the preparation of thermoplastic blends based on elastomers and plastomers, such as, for instance, Banbury-type internal mixers, Buss-type continuous masticators/extruders, two-screw corotating or counterrotating masticating drawplates, two-roller calenders, or the like. To the homogeneous mass so obtained, still moving and brought to

a temperature comprised between 160 and 230°C, preferably between 180 and 210°C, the two chemical compositions are added which constitute the vulcanization system and the mass is kept in movement until the wished degree of vulcanization of the elastomer is reached, which preferably should not exceed 95% in weight. The binary vulcanization system, consisting of a mix of an alkylphenol-formaldehyde and sodium bisulphite mix, is added in such an amount as to cause the alkylphenol-formaldehydic resin to be comprised between 2 and 20, preferably between 3 and 15, weight parts per 100 weight parts of the elastomer.

In the binary vulcanization system, sodium bisulphite varies from 50 and 150, preferably from 60 to 130, weight parts per 100 weight parts of the alkylphenol-formaldehydic resin; as a consequence, the weight ratio between sodium bisulphite and alkylphenol-formaldehydic resin is comprised between 5:10 and 150:100.

The time necessary for the vulcanization varies according to the temperature variation and is generally comprised between 30 seconds and 30 minutes.

By way of example and orientation, some times of vulcanization in function of temperature are given hereunder:

at 160°C: 10 - 30 minutes,

at 170°C: 5 - 10 minutes,

at 180°C: 1 - 5 minutes,

at 190°C: 60 seconds - 2 minutes,

at 200°C: 30-60 seconds.

Having completed the vulcanization, the plastoelastomeric composition so obtained is cooled and transformed by extrusion in

a physical form (generally granules or pellets) suitable for an easy utilization with the techniques usually utilized for thermoplastic materials, such as, for instance, drawing, moulding, calendering, etc.

The plastoelastomeric compositions so obtained have improved processeability and elasticity properties, and, in general, improved-physical mechanical characteristics compared to the known plastoelastomeric compositions.

#### Elastomeric Phase

The elastomers utilized as elastomeric phase in the composition subject matter of this invention are the EPDM ones known for their excellent vulcanizing properties, such as resistance to ozone, protracted heat, oxygen, UV radiation, basic and acid chemical agents.

EPDM elastomers are structurally amorphous polymers constituted by ethylene, by at least another alpha-olefin having a higher number of carbon atoms, for instance from 2 to 6, such as propylene or butylene-1, and by at least a dienic monomer, which has preferably the non-conjugated configuration. For this reason EPDM elastomers are defined as saturated polymeric chain polymers, having therefore a high chemical inertia.

Examples of dienic monomers include:

1,4-hexadiene, 1,5-hexadiene, 1,5-cyclooctadiene, 2-methyl-1,4-pentadiene, dicyclopentadiene, ethylidene-norbornene, butadiene or associated mixes, and preferably 1,4-hexadiene, dicyclopentadiene, ethylidene-norbornene or associated mixes.

Preferred EPDM elastomers are those obtained by polymerization of

monomers by metallorganic catalysis and vanadium soluble salts and have a molecular weight comprised between 100,000 and 700,000, an ethylene contents comprised between 20 and 80% in moles, a bound dienic monomer content comprised between 3 and 18% in weight per 100 parts in weight of polymer, the rest being propylene.

#### **Plastomeric Phase**

Thermoplastic olefinic polymers which form the plastomeric phase of plastoelastomeric compositions subject matter of this invention are homopolymers or copolymers having a high molecular weight (comprised between 100 and 100,000) obtained from the polymerization of  $C_2-C_8$  alpha-olefins, such as ethylene, propylene, butene-1, pentene-1, 1-methylpentene-1, 2-methylpentene-1, 3-methylpentene-1, 4-methylpentene-1, hexene-1, 1-methylhexene-1, 2-methylhexene-1, 3-methylhexene-1, 4-methylhexene-1, 5-methylhexene-1, heptene-1.

Preferably one or more olefinic polymers of the above mentioned type, and preferably polyethylene, of either the high-density or the low-density or the linear type, and/or isotactic or syndiotactic cristalline polypropilene and/or propylene-ethylene copolymers, are preferably used to the purposes of this invention.

#### **Elastomer/Plastomer Mixes**

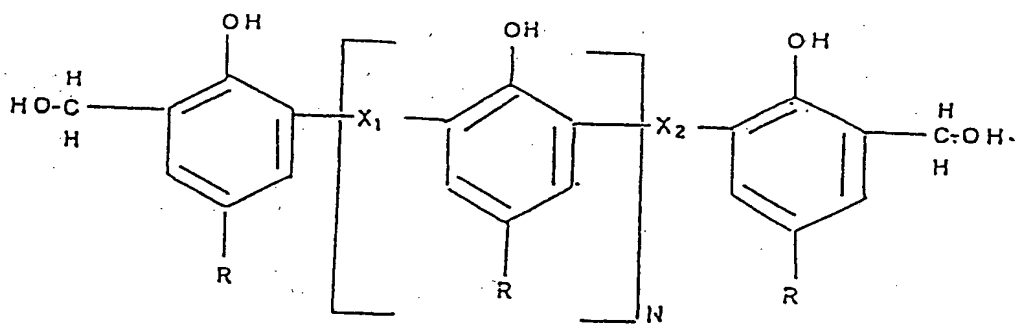
According to this invention, the EPDM elastomer/polyolefinic plastomer mixes are constituted by weight ratios that vary according to the final use of the compositions and they may vary from 10 to 90% in weight of elastomer and from 90 to 10% in weight of plastomer; preferably from 60 to 85% in weight of

elastomer and from 40 to 15% in weight of plastomer.

#### Vulcanization System

The vulcanization system of this invention is an alkylphenol-formaldehydic resin utilized with sodium bisulphite. As yet, this binary system has neither been utilized for the vulcanization of dienic rubber in general nor for EPDM rubber in particular. Surprisingly, the applicant has now discovered that its utilization in the dynamic vulcanization of EPDM elastomeric phase of plastoelastomeric compositions lends the final compositions better characteristics of processability, elasticity and generally better physical-mechanical properties compared to those obtained by analogous compositions which carry out dynamic vulcanization by means of other known vulcanization systems (vulcanization agents and coagents). Alkylphenol-formaldehydic resins are known as vulcanization agents of low dienic content-rubbers, such as, for instance, butylic rubber (generally referred to as I I R, i.e. Isoprene-Isobutene Rubbers), but only in combination with donors of halogens or together with metal oxides (see Werner Hofman, "Vulcanization and Vulcanizing Agents", pp. 300-301, MacLaren and Sons Ltd., London), according to reaction schemes widely illustrated in said text.

The alkylphenol-formaldehydic resin utilized together with sodium bisulphite for the dynamic vulcanization of the EPDM elastomeric phase of the plastoelastomeric composition subject matter of this invention has the following general formula:



where:

- $X_1$  and  $X_2$  are radicals -  $\text{CH}_2$  - or  $\text{CH}_2\text{-O-CH}_2$ ;
- R is an alkylic radical containing 4 to 16 carbon atoms;
- N is nought or a whole number between 1 and 6.

These compositions are prepared by condensation of alkylphenols with formaldehyde, according to known techniques.

By the term "sodium bisulphite" one means the sodium salt of sulphurous acid. This composition is also called "acid sodium sulphite" and corresponds to the following general formula:  
 $\text{NaHSO}_3$ .

This composition is widely illustrated in the literature and is mentioned as a reducing agent due to the easiness it oxidizes into sulphate.

The process to obtain the plastoelastomeric compositions subject matter of this invention comprises the intimate mixing at the molten state of an olefinic plastomer with an EPDM elastomer composed by ethylene, another alpha-olefinic monomer having a number of carbon atoms greater than 2 and a dienic monomer as illustrated above. Such intimate mixing is obtained by submitting to mastication the components that form the mix, at a suitable temperature and according to weight ratios from 10 to 90% of plastomer and from 90 to 10% of EPDM elastomer, the binary vulcanization system being constituted by an alkylphenol-formaldehydic resins whose amount is comprised between 2 and 20% in weight compared to EPDM elastomer, and by sodium bisulphite, whose amount is comprised between 50 and 150 in weight compared to said alkylphenol-formaldehydic resin.

Mastication is carried on until the elastomer has vulcanized,



which requires a variable time depending on work temperature, as previously illustrated, and with a vulcanization degree which may reach 95% in weight.

The elastomer, dynamically vulcanized in this way, is finely and homogeneously dispersed in the plastomer, in form of very small particles of the order of the micron. The smallness of particles will be directly proportional to the shear stress of the rotatory apparatus utilized.

The plastoelastomeric compositions subject matter of this invention may include additives that are usually utilized in rubber and plastic material industries, such as pigments, organic and inorganic dyes, plasticizers such as paraffin and naphthene oils, fatty acids, fatty acid salts, organic extenders, such as, for instance, channel or funnel process carbon black, inorganic extenders, such as, for instance, carbonates, silicates, kaolins, talc and clays, and also agents stabilizing against heat, oxidation, ageing, etc.

The compositions subject matter of this invention can be transformed into artifacts or bodies formed by injection and/or compression moulding, calandering, extrusion, blow-moulding or rotomoulding, according to known techniques.

The following examples are given to the purpose of better illustrating this invention and should not be construed as limiting same.

#### Examples 1-9

Example 1-9 stress the high physical-mechanical, elasticity and processability properties of the plastoelastomeric compositions

obtained according to this invention, by utilizing different commercial types of EPDM elastomer and of polypropylenic olefinic plastomer, with a different weight ratio between the two of them. An homogeneous mix of an elastomer of the type and in the quantity as specified on Table 1 and a polypropylene of the type and in the quantity as specified on Table 1 was prepared in a Banbury-type internal mixer. The operating features of the mixer were:

- Temperature of the machine: 180°C
- Speed of rotors: 90 revolutions/minute
- Nominal capacity of the machine: 22 liters
- Filling coefficient: 1,15
- Mixing time: 2 minutes
- Mass temperature: 190°C.

The binary vulcanization system and stearic acid were then added to the moving mass, in the quantities specified on Table 1. After 2 minutes and 30 seconds, an 8% increase in mass temperature and a 7% increase in electric energy consumption, detected by the connected amperometer, were observed: these changes point out to the vulcanization being completed. The unloaded and cooled mass was then moulded in a Triulzi-type 300 g injection mould for plastic materials, and then characterized.

For the determination of the characteristics of the compositions obtained from both these and the subsequent examples, the following analysis methods were followed:

- Shore A and Shore D hardness : ASTM D2240
- Ultimate tensile stress: ASTM D412
- 100% elongation modulus: ASTM D412

- 200% elongation modulus: ASTM D412
- Ultimate elongation: ASTM D412
- Tearing resistance: ASTM D624
- 100% elongation tension: ASTM D412
- 25% deflection: ASTM D395/B

Processeability was determined by measuring head pressure with extrusion tests in laboratory drawplate, the operating features being as follows:

- Temperature of die chaser: 195°C
- Temperature of machine body: 180°C
- Diameter of die chaser: 3 mm
- Length/diameter ratio (L/D): 28
- Diameter of screw (D): 20 mm
- Delivery: variable, depending on processeability degree
- Head pressure: variable, depending on processeability degree

The percent of vulcanized EPDM was determined by solubility in xylol on boiling: quantities are always referred to 100 parts in weight of pure elastomer, without taking into account the extension oil possibly included in the base polymer.

Table 1 shows the compositions expressed in weight parts and the characteristics of same.

T A B L E 1									
EXAMPLES	1	2	3	4	5	6	7	8	9
EPDM (°)	130	130	130						
EPDM (°°)				125	125	125			
EPDM (°°°)							200	200	200

Polypropylene (i)	40	45	65						
Polypropylene (ii)				40	50	65			
Polypropylene (iii)							40	50	65
Paraffinic oil	150	90	70	160	85	85	70	10	10
Stearic acid	2	2	2	2	2	2	2	2	2
Sodium bisulphite	6	6	6	9	9	9	9,5	9,5	9,5
Alkylphenol-									
formaldehydic resin	8	8	8	10	10	10	11	11	11

#### Characteristics of the compositions

##### Shore A hardness

points	55	65	75	60	70	75	60	70	75
U.T.S. kg/cm <sup>2</sup>	60	70	85	70	85	90	65	75	85
100% modulus kg/cm <sup>2</sup>	15	20	31	18	20	32	20	25	30
200% modulus kg/cm <sup>2</sup>	25	32	41	38	45	48	35	40	45
% ultimate elongat.	600	620	650	630	650	680	610	630	650

##### Tearing resistance

kg/cm	25	28	30	25	29	35	27	30	36
100% tension, %	10	12	13	11	12	13	10	11	13
25% deflection									
(72 h, 70°C), %	31	33	35	32	33	36	31	32	34

#### Die drawing tests

Delivery g/min	50	45	48	45	50	55	50	52	58
Head pressure kg/cm <sup>2</sup>	250	240	190	220	190	170	190	170	165
Vulcanized EPDM									

% weight

93

94

93

94

94

94

95

95

95

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**Description of ingredients utilized**

**EPDM (°):** Vistalon 6630, produced and marketed by EXXON Chem., having the following composition: propylene = 30% in weight, ethylidene-norbornene non saturated monomer = 8% in weight; paraffinic oil = 23% in weight; Mooney viscosity, ML 1+4 at 121°C = 40.

**EPDM (°°):** Dutral TER 235/E2, produced and marketed by Montedison S.p.A., having the following composition: propylene = 32% in weight; ethylidene-norbornene non saturated monomer = 7,5% in weight; paraffinic oil = 20% in weight; Mooney viscosity, ML 1+4 at 121°C = 40.

**EPDM (°°°):** Keltan 509X100, produced and marketed by D S M - Holland, having the following composition: propylene = 27% in weight; ethylidene-norbornene non saturated monomer = 7% in weight; Mooney viscosity, ML 1+4 at 121°C = 50.

**Polypropylene (i):** Moplen D 50 S, produced and marketed by Himont S.p.A., having a flowability degree at 230°C with a weight of 21,6 Newton = 0,3 g/10 min.

**Polypropylene (ii):** Laqtene P.3020 GN3, produced and sold by ATO Chemie (France), having a flowability degree at 230°C with a weight of 21,6 Newton = 1,8 g/10 min and ethylene contents = 3,5% in weight and propylene contents = 96,5% in weight.

**Polypropylene (iii):** Novolen 1300 E, produced and sold by BASF (Germany), having a flowability degree at 230°C with a weight of 21,6 Newton = 0,4 g/10 min.

Paraffinic oil: Primol 352, produced and sold by EXXON Chem.

Alkylphenol-formaldehydic resin: SP 1045, produced and sold by Schenectady Chemicals, Inc., Midland (US).

#### Examples 10-13

The following examples stress the better processeability as well as the better elastic properties of the plastoelatomeric compositions subject matter of this invention (example 10), compared to those (example 11-13) obtained with other known vulcanizing systems.

The compositions shown on Table 2 were fed, in the quantities given in such Table, to a compounder/extruder of the Baker-Perkins type, with diameter = 50 mm, Length/Diameter (L/D) ratio = 25, equipped with gravimetric batcher for solids and a piston batcher for the plasticizing oil. An one-screw side extruder permits, by head-cutting in water, to give the compositions the physical form of pellets. The EPDM elastomer had been previously broken into small irregular particles by means of a cutter, in order to render it feedable by means of a weight-batcher.

Operating temperatures:

Zone B: 190°C; Zone C: 190°C; Zone D = 200°C; Zone E = 205°C; Zone F = 210°C; Zone G = 220°C; Zone H = 225°C; Zone J = 228°C; Zone K = 230°C.

Die chaser of the one-screw side extruder = 200°C; optimal number of revolutions = 230/min; recorded head pressure = 80 bar; delivery per hour = 65 kg.

The EPDM elastomer was introduced by means of weight-batcher nr. 1, the polypropylene plastomer by means of batcher nr. 2, and the other ingredients - dry-premixed by special turbomixer - by means

of batcher nr. 3 (batcher for small weight quantities).

The plasticizing oil was introduced into the coumpounder-extruder by means of piston-batcher preheated at 120°C.

The analyses of the compositions obtained were carried out according to the same method utilized for the preceding examples.

EPDM rubber is the Vistalon 6630 indicated by (°) in example 1, polypropylene is the Moplen D 50 S indicated by (i) in the preceding example, and the alkylphenol-formaldehydic resin is the S P type produced and sold by company Schenectady.

Table 2 shows the compositions expressed in weight parts and the main characteristics of same. The composition of example 12 shows clear signs of degradation of polypropylene.

T A B L E 2

EXAMPLES	10	11*	12*	13*
EPDM (°)	130	130	130	130
Polypropylene (i)	40	40	40	40
Paraffinic oil	110	110	110	110
Stearic acid	2	2	2	2
Vulcanization system:				
- A	20	-	-	-
- B	-	22	-	-
- C	-	-	9,8	-
- D	-	-	-	20

Characteristics of the compositions

Shore A hardness, points	60	60	54	62
U.T.S., kg/cm <sup>2</sup>	75	61	48	58
100% modulus, kg/cm <sup>2</sup>	18	13	9	12
200% modulus, kg/cm <sup>2</sup>	32	28	12	25
% ultimate elongation	620	600	860	650
Tearing resistance, kg/cm	45	33	18	32
100% tension, %	10	15	33	18
25% deflection (72 h at 70°C), %	28	32	45	36
Extrusion tests:				
Delivery, g/min	45	28	40	30
Head pressure, kg/cm <sup>2</sup>	220	250	120	250
Vulcanized EPDM, weight %	95	98	99	99

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\* Comparison examples

Description of the vulcanization systems	-	Parts in weight
- A		
Alkylphenol-formaldehydic resin SP 1045		7
Sodium bisulphite		7
Total		14
- B		
Bromomethyl-alkylphenolic resin containing		
6% in weight of halogen and carrying the		
trade mark SP 1055, produced and sold by		
company Schemectady Chemicals, Inc.,		
Midland (US)		6
Active zinc oxide		5
Total		11



- C

Inert-carried dicumene peroxide with active agent = 40% in weight, carrying the trade mark Peroximon D C 40 and produced by company Montefluos (Italy)	4,5
Sulphur	0,3
Active zinc oxide	5
Total	9,8

- D

Alkylphenol-formaldehydic resin S P 1045	10
Magnesium oxide	8
Total	18

#### Examples 14-19

These examples stress the efficaciousness and the synergism of the binary vulcanization system of this invention in the production of plastoelastomeric compositions having a dynamically vulcanized elastomeric phase, in terms of physical-mechanical and elasticity properties.

In these examples, the same operating methods were followed as in examples 10-13.

Table 3 shows the compositions expressed in weight parts and the main characteristics of same, determined according to the standard methods described for example 1.

#### Description of ingredients utilized:

EPDM: the one marked by (°°°) in examples 7-9, i.e. Keltan 509X100.

Polypropylene: the one marked by (i) in examples 1-3, i.e. Moplen D 50 S.

**Polyethylene:** Eraclene H ZB 6015; high density homopolymer produced by Enichem Polimeri (Italy), having density = 0,960 g/cm<sup>3</sup>; a flowability degree at 190°C (with weight of 2,16 Newton) = 0,30 g/10 minutes.

**Alkylphenol-formaldehydic resin:** the same of examples 1-9. i.e. S P 1045.

**Paraffinic oil:** trade name Paraffinic Oil O B 10, produced by AGIP Petroli (Italy).

T A B L E 3

EXAMPLES	14*	15*	16	17*	18*	19
EPDM (°°°)	200	200	200	200	200	200
Polypropylene (i)	40	40	40	-	-	-
Polyethylene	-	-	-	50	50	50
Stearic acid	2	2	2	2	2	2
Paraffinic oil	40	40	40	30	30	30
Sodium bisulphite	-	-	10	-	-	10
SP 1045 resin	-	10	10	-	10	10

Characteristics of the compositions

Shore A hardness, points	62	64	65	65	67	72
U.T.S., kg/cm <sup>2</sup>	35	50	75	30	44	80
100% modulus, kg/cm <sup>2</sup>	9	11	26	7	12	33
200% modulus, kg/cm <sup>2</sup>	20	27	38	18	23	42
Ultimate elongation, %	900	890	600	950	840	600
Tearing resistance, kg/cm	30	28	35	32	35	43

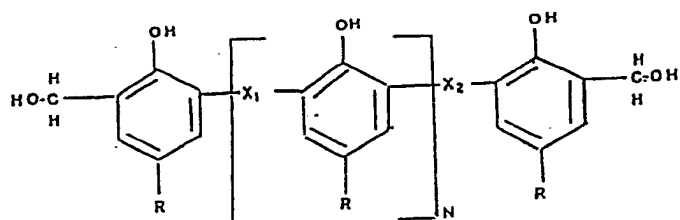
100% tension, %	90	62	12	82	55	14
25% deflection (72 h						
at 70°C, %	92	69	32	88	65	31
Die drawing test:						
Delivery, g/min	100	80	40	120	100	75
Head pressure, kg/cm <sup>2</sup>	120	180	220	90	120	200
Vulcanized EPDM, % weight	0	45	95	0	55	94,6

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\* Comparison examples

# CLAIMS

1. Process for the preparation of plastoelastomeric compositions composed by an elastomeric phase constituted by EPDM elastomers and by a plastomeric phase constituted by thermoplastic olefinic polymers, wherein the elastomeric phase is dynamically vulcanized by means of a vulcanizing agent, characterized in that the vulcanizing agent is a binary system consisting of an alkylphenol-formaldehydic resin and sodium bisulphite  $\text{NaHSO}_3$ .
2. Process according to claim 1, characterized in that the alkylphenol-formaldehydic resin has the general formula



where:

- $X_1$  and  $X_2$  are radicals -  $\text{CH}_2$  - or  $\text{CH}_2\text{-O-CH}_2$ ;
  - R is an alkylic radical containing 4 to 16 carbone atoms; and
  - N is nought or a whole number from 1 to 6.
3. Process according to claim 1 or 2, characterized in that the vulcanization degree of the elastomeric phase is not greater than 95% of the total initial weight of said elastomeric phase.
  4. Process according to any of the preceding claim, characterized in that the elastomeric phase varies from 10 to 90 parts and, correspondingly, the plastomeric phase varies from 90 to 10 parts per 100 parts of elastomer/plastomer mix.
  5. Process according to any of the claims from 1 to 4, characterized in that the alkylphenol-formaldehydic resin is utilized in weight amounts variable from 2 to 20, preferably from 3 to 15, parts in weight per 100 parts in weight of elastomeric

phase.

6. Process according to any of claims from 1 to 5, characterized in that sodium bisulphite is utilized in weight amounts variable from 50 to 150, preferably from 60 to 130, parts in weight per 100 parts in weight of alkylphenol-formaldehyde resin.
7. Process according to any of claims from 1 to 6, characterized in that the elastomeric phase is an EPDM elastomer constituted by ethylene, another alpha-olefin, having a greater number of carbon atoms, and a dienic monomer such as 1,4-hexadiene, 1,5-hexadiene, 1,5-cyclooctadiene, 2-methyl-1,4-pentadiene, dicyclopentadiene, ethylidene-norbornene and butadiene.
8. Process according to claim 7, wherein the EPDM elastomer has a molecular weight comprised between 100,000 and 700,000, an ethylene contents comprised between 20 and 80% in moles, a dienic monomer contents from 3 to 18% in weight, the rest being constituted by propylene.
9. Process according to any of claims from 1 to 8, characterized in that the polyolefinic plastomeric phase consists of one or more homopolymers, or copolymers of alpha-olefins such as ethylene, propylene, butene-1, pentene-1, 1-methylpentene-1, methylpentene-1, 3-methylpentene-1, 4-methylpentene-1, hexene-1, 1-methylhexene-1, 2-methylhexene-1, 3-methylhexene-1, 4-methylhexene-1, 5-methylhexene-1, heptene-1.
10. Process according to claim 9, wherein the polyolefinic plastomeric phase is chosen among polyethylene, either of the high-density or the low-density type or the linear type, polypropylene, either of the isotactic or the syndiotactic type, and propylene-ethylene copolymers.

11. Plastoelastomeric composition composed by an elastomeric phase constituted by EPDM elastomers and by a plastomeric phase constituted by thermoplastic olefinic polymers, wherein the elastomeric phase is dynamically vulcanized in presence of a binary vulcanizing system consisting of an alkylphenol-formaldehydic resin and sodium bisulphite.

12. Artifacts obtainable from the plastoelastomeric composition of claim 9, by injection and/or compression moulding, calendering, extrusion, blow-moulding, or rotomoulding.

13. Process for the preparation of plastoelastomeric compositions substantially as herein described.

14. Plastoelastomeric composition according to Claim 11 substantially as herein described.

15. Artifacts according to Claim 12 substantially as herein described.

**Patents Act 1977**

**Examiner's report to the Comptroller under Section 17  
(The Search report)**

22  
Application number  
GB 9324361.6

**Relevant Technical Fields**

- (i) UK Cl (Ed.M) C3J JAB  
(ii) Int Cl (Ed.5) C08J, C08K, C08L

Search Examiner  
M J PRICE

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17 FEBRUARY 1994

**Databases (see below)**

(i) UK Patent Office collections of GB, EP, WO and US patent specifications.

Documents considered relevant  
following a search in respect of  
Claims :-  
1-15

(ii) ONLINE DATABASES: WPI

**Categories of documents**

- X: Document indicating lack of novelty or of inventive step. P: Document published on or after the declared priority date but before the filing date of the present application.  
Y: Document indicating lack of inventive step if combined with one or more other documents of the same category. E: Patent document published on or after, but with priority date earlier than, the filing date of the present application.  
A: Document indicating technological background and/or state of the art. &: Member of the same patent family; corresponding document.

Category	Identity of document and relevant passages		Relevant to claim(s)
A	EP 0270020	(POLYSAR) see eg Claim 1	
A	EP 0213285	(HIMONT) see eg Claim 1	
A	EP 0046285	(MONTEDISON) see eg Claim 1	
A	US 4835204	(SOFTER) see eg Claim 1	

**Databases:** The UK Patent Office database comprises classified collections of GB, EP, WO and US patent specifications as outlined periodically in the Official Journal (Patents). The on-line databases considered for search are also listed periodically in the Official Journal (Patents).

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